Staggered-spin contribution to nuclear spin-lattice relaxation in two-leg antiferromagnetic spin-1/2 ladders.

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We study the nuclear spin-lattice relaxation rate $1/T_1$ in the two-leg antiferromagnetic spin-1/2 Heisenberg ladder. More specifically, we consider the contribution to $1/T_1$ from the processes with momentum transfer (π,π). In the limit of weak coupling between the two chains, this contribution is of activation type with gap 2Δ at low temperatures (Δ is the spin gap), but crosses over to a slowly-decaying temperature dependence at the crossover temperature $T \approx \Delta$. This crossover possibly explains the recent high-temperature NMR results on ladder-containing cuprates by T. Imai et al.

Recent NMR experiments on Cu_2O_3 ladders in $A_{14}Cu_{24}O_{41}$ compounds (A_{14}=La_6Ca_8, Sr_{14}, $Sr_{11}Ca_3$) reveal unexpected behavior of the nuclear spin-lattice relaxation rate $1/T_1$ at temperatures of order of the spin gap [1]. While both $^{63}1/T_1$ and $^{17}1/T_1$ exhibit activation-type behavior at low temperature, a well-pronounced crossover is observed in ^{63}Cu nuclear spin relaxation rate. There exist numerous theoretical studies of $1/T_1$ in antiferromagnetic two-leg ladders [2–5], but none of them predicts a similar crossover. In this paper we restrict our attention to the undoped spin ladder and argue that the observed crossover is due to the processes with the momentum transfer $q = (\pi,\pi)$.

We describe the spin ladder by the Heisenberg Hamiltonian

$$H = \sum_n \left[J(\vec{S}_n^{(I)} \vec{S}_{n+1}^{(I)} + \vec{S}_n^{(II)} \vec{S}_{n+1}^{(II)}) + J_\perp \vec{S}_n^{(I)} \vec{S}_n^{(II)} \right]. \tag{1}$$

Here n is an integer labeling the rungs of the ladder, the superscripts refer to the two chains. J and $J_\perp$ are the coupling constants. From analyzing experimental data on magnetic susceptibility [6] and on ^{17}O NMR Knight shift [1], it has been suggested that in Cu_2O_3 ladders $J_\perp/J \approx 0.5$.

Both the strong-coupling ($J_\perp/J \gg 1$) and the weak-coupling ($J_\perp/J \ll 1$) approaches confirm that the low-lying magnetic excitations in the ladder are spin-1 magnons with the minimal gap at $k = (\pi,\pi)$; in the physically relevant range $J_\perp/J \approx 0.5$, the value of the gap is known to be $\Delta \approx 0.5 J_\perp$ [7,5,8]. We believe that the weak-coupling limit captures the characteristic features of the nuclear spin relaxation. For the sake of simplicity we assume that $T, J_\perp \ll J$ (with no assumptions on the relative magnitudes of $J_\perp$ and the temperature T). Then all excited magnons have momenta close to the wavevector $k = (\pi,\pi)$. The relaxation rate $1/T_1$ is given by

$$\frac{1}{T_1} = 2\sum_q F(q) S(q,\omega_0) = \sum_q F(q) \frac{2T}{\omega_0} \mathrm{Im}\chi(q,\omega_0), \tag{2}$$

where $F(q)$ are the appropriate coupling constants, ω_0 is the nuclear resonance frequency ($\omega_0 \ll \Delta, T$), $S(q,\omega)$ is the dynamical structure factor, and $\chi(q,\omega)$ is the dynamical magnetic susceptibility. We employ the system of units with $\hbar = k_B = \mu_B = 1$. Since the excited magnons have momenta close to (π,π), there are two major contributions to $1/T_1$: that with $q \approx (0,0)$ (via even-magnon-number processes) and that with $q \approx (\pi,\pi)$ (via odd-magnon-number processes). Accordingly, define

$$\left(\frac{1}{T_1}\right)_{q=0} = 2\int_{q\approx 0} \frac{dq}{2\pi} S((q,0),\omega_0), \qquad \left(\frac{1}{T_1}\right)_{q=\pi} = 2\int_{q\approx\pi} \frac{dq}{2\pi} S((q,\pi),\omega_0) \tag{3}$$

(defined this way, the quantities $(1/T_1)_{q=0}$ and $(1/T_1)_{q=\pi}$ do not have the dimension of inverse time). Then the total relaxation rate $1/T_1$ is a linear combination of $(1/T_1)_{q=0}$ and $(1/T_1)_{q=\pi}$.

The coupling constants $F(q)$ for the oxygen sites vanish at $q = (\pi,\pi)$ [1], so

$$^{17}\frac{1}{T_1} \propto \left(\frac{1}{T_1}\right)_{q=0}. \tag{4}$$

The experiments on $SrCu_2O_3$ suggest that in Cu_2O_3 ladders the coupling constant $F(q)$ for the copper site is dominated by the on-site hyperfine interaction and therefore only weakly depends on q [9]. Thus we expect that $^{63}1/T_1$ is a linear combination of $(1/T_1)_{q=0}$ and $(1/T_1)_{q=\pi}$ with the coefficients of the same order of magnitude. Next, the available numerical studies [4] indicate that at temperatures of order of the spin gap the contributions $(1/T_1)_{q=0}$ and $(1/T_1)_{q=\pi}$

have comparable magnitudes. Therefore, we suggest that $^{63}1/T_1$ in the experimentally relevant temperature range is not dominated by the $q=0$ contribution, in contrast with other studies [2,3].

A simple argument shows that the contribution $(1/T_1)_{q=\pi}$ has gap 2Δ as opposed to gap Δ for $(1/T_1)_{q=0}$ (this argument was originally proposed for spin-1 chains [10], but it also remains valid for spin-1/2 ladders). In $(1/T_1)_{q=0}$, the nuclear spin is relaxed by the quasi-elastic scattering of a thermally excited magnon, which requires a gap of Δ. On the other hand, $(1/T_1)_{q=\pi}$ assumes an elastic scattering with momentum transfer near (π,π), which means that the total number of incoming and outcoming magnons must be odd (each magnon carries momentum close to (π,π)). Processes of creating and annihilating a single magnon cannot occur at zero energy because of the gap. Therefore, the leading contribution comes from three-magnon processes: two thermally excited magnons are converted into a single magnon carrying the total energy of the incoming magnons (up to the NMR frequency ω_0), or vice versa (see Fig. 1 below). Such processes require energy at least 2Δ.

On the basis of the above argument, the contribution $(1/T_1)_{q=\pi}$ was usually neglected. However, several recent works conclude that the effective gap in $(1/T_1)_{q=0}$ is somewhat larger than Δ (determined from succeptibility), either because of magnon interaction [3] or due to the singlet excitation mode [2]. A larger gap in $1/T_1$ than in the succeptibility is indeed reported in Cu_2O_3 ladders [11,9]. In view of these results, our suggestion of importance of $q=\pi$ contribution to ^{63}Cu nuclear spin relaxation rate appears more plausible.

For the rest of the paper we focus on computing $(1/T_1)_{q=\pi}$ in the weak-coupling limit $J_\perp \ll J$. Although we are unable to find a closed analytic form for $(1/T_1)_{q=\pi}$ in the whole range of temperatures, we find the high- and low-temperature asymptotics and estimate the crossover temperature.

According to the results of [8], in the weak-coupling limit, the spin-1/2 two-leg ladder is equivalent to four massive Majorana fermions, combined into a triplet of mass Δ and a singlet of mass 3Δ:

$$H_f = H_\Delta[\xi_1] + H_\Delta[\xi_2] + H_\Delta[\xi_3] + H_{3\Delta}[\rho] + H_{int}, \tag{5}$$

where each $H_m[\xi]$ is a free massive Hamiltonian of a Majorana fermion ξ; H_{int} is the four-fermion interaction arising from the marginal term in the interchain coupling [8]. It has been argued in [3] that the interaction would lead to nonperturbative effects in $(1/T_1)_{q=0}$. However, the staggered magnetization is nonlocal in terms of fermions, and we expect that the interaction will play a smaller role in $(1/T_1)_{q=\pi}$. Following [8], we neglect H_{int}.

In the continuum limit, the local magnetization $\vec{S}^{(\alpha)}$ may be split into the uniform and staggered components:

$$\vec{S}^{(\alpha)}_n = \vec{J}^{(\alpha)} + (-1)^n \vec{n}^{(\alpha)}. \tag{6}$$

While $\vec{J}$ has a quadratic expression in terms of the Majorana fermions, $\vec{n}$ is non-local in the fermion operators. If the Majorana fermions are mapped onto non-critical 1+1-dimensional Ising models (we need four Ising models — one for each Majorana fermion), the operator $\vec{n}$ may be expressed in terms of order (σ) and disorder (μ) parameters of the Ising models [8]. In particular,

$$n_z^- = n_z^{(I)} - n_z^{(II)} \propto \sigma_1\sigma_2\mu_3\sigma^*, \tag{7}$$

where σ^* is the order parameter for the Ising model with gap $\Delta^* = 3\Delta$; σ_1, σ_2, and μ_3 — order and disorder parameters of the three identical Ising models with gap Δ [8]. Conventionally, we have chosen the Ising models to be in ordered state, so that $\langle\sigma\rangle \neq 0$ at zero temperature.

The expression (7) would enable us to compute $(1/T_1)_{q=\pi}$ using (3), should we know the non-critical Ising correlation functions at finite temperature. In a recent work [12], the latter has been expressed as a series in number of fermionic excitations, and we shall use their result to match the high- and low-temperature asymptotics of the Ising correlation functions.

The prportionality coefficient in (7) is non-universal. To fix the relative normalization of the high- and low-temperature asymptotics, define

$$\tilde{n}_z^- = \sigma_1\sigma_2\mu_3\sigma^* \tag{8}$$

with the order and disorder operators normalized by their short-distance asymptotics:

$$\langle\sigma(x)\sigma(0)\rangle \sim \langle\mu(x)\mu(0)\rangle \sim \frac{1}{|x|^{1/4}}, \qquad x\to 0. \tag{9}$$

In what follows we normalize $(1/T_1)_{q=\pi}$ accordingly, in other words, in (3) we set $S(x,t) = \langle\tilde{n}_z^-(x,t)\tilde{n}_z^-(0,0)\rangle$.

In a natural way, the following three limits may be distinguished:

(i) $T \ll \Delta$. In this low-temperature limit, $(1/T_1)_{q=\pi}$ has activation-type behavior with gap 2Δ. The prefactor may be computed using the results of [12].

(ii) $T \gg \Delta^*$. In this limit the gaps presumably play no role, and the system is equivalent to two uncoupled chains ("quantum critical" phase). The result for $(1/T_1)_{q=\pi}$ in this limit may be borrowed from [13]. Although this high temperatures are beyond the experimental range, considering this limit is useful for determining the degree of applicability of the approximations being made.

(iii) $\Delta \ll T \ll \Delta^*$. In the ladder system this intermediate limit is never attained, since $\Delta^* = 3\Delta$. However we may formally take this limit assuming $\Delta^* \gg \Delta$. The validity of this approximation will be discussed further.

Below we describe in more detail the computations and the results in these three limits.

(i) $T \ll \Delta$. According to [12], the (non-ordered) Ising correlation functions may be expressed as the series in the number of fermionic excitations:

$$\langle \sigma(t,0)\sigma(0,0)\rangle = \sigma_0^2 \sum_{n\ \mathrm{even}} \frac{1}{n!} \sum_{\varepsilon_i} \int_{-\infty}^{+\infty} \prod_{i=1}^{n} \frac{d\beta_i}{2\pi} f_{\varepsilon_i}(\beta_i) e^{-i\varepsilon_i E(\beta_i)t} |F(\beta_1,\ldots,\beta_n)_{\varepsilon_1,\ldots,\varepsilon_n}|^2, \tag{10}$$

where $\varepsilon_i = \pm 1$ $(i = 1,\ldots,n)$ are the particle-hole indices,

$$f_\varepsilon(\beta) = \left[1 + \exp(-\varepsilon E(\beta)/T)\right]^{-1} \tag{11}$$

is the Fermi distribution function, β_i are the rapidities of the excitations with energies $E(\beta) = \Delta \cosh \beta$.

$$F(\beta_1,\ldots,\beta_n)_{\varepsilon_1,\ldots,\varepsilon_n} = i^{n/2} \prod_{i<j} \left(\tanh \frac{\beta_i - \beta_j}{2}\right)^{\varepsilon_i \varepsilon_j} \tag{12}$$

are the corresponding formfactors [12,14].

The same expression gives $\langle \mu(t,0)\mu(0,0)\rangle$, with the only difference that the sum is taken over odd numbers of excitations n.

The zero-temperature magnetization σ_0 may be taken from the exact result on the Ising model [15,16]:

$$\sigma_0^2 = \Delta^{1/4} 2^{1/6} e^{-1/4} A^3 \approx 1.8437 \Delta^{1/4}, \tag{13}$$

where $A = \exp[1/12 - \zeta'(-1)] = 1.282427\ldots$ is the Glaisher constant. In the Ising model with gap 3Δ, ${\sigma_0^*}^2 = 3^{1/4}\sigma_0^2$.

Using (10), we express

$$\left(\frac{1}{T_1}\right)_{q=\pi} = 2 \int_{-\infty}^{+\infty} dt \langle \tilde{n}_z^-(x,t) \tilde{n}_z^-(0,0)\rangle \tag{14}$$

as a sum of the odd-magnon-number processes with zero energy transfer. The one-magnon process has a gap and does not contribute to (14). In Fig. 1 we show the three types of three-magnon processes contributing to $(1/T_1)_{q=\pi}$.

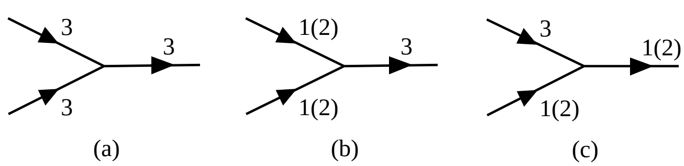

FIG. 1. Three-magnon processes contributing to $(1/T_1)_{q=\pi}$.

In the diagrams, the arrows correspond to the particle-hole index ε_i in (10), the labels 1,2,3 — to the three different Majorana fermions (equivalently, to the spin of magnons). The contribution from the diagrams shown in Fig. 1 must be multiplied by two to account for the diagrams with arrows reversed. We neglect processes including the singlet channel (with gap 3Δ), since they have a larger gap 4Δ. The processes (a) and (b) in Fig. 1 can be shown to have temperature dependence $T^2 \exp(-2\Delta/T)$, while the process (c) dominates at low temperatures with the temperature dependence $T \exp(-2\Delta/T)$. Including only the latter contribution, we find:

$$\left(\frac{1}{T_1}\right)_{q=\pi} = 4\sigma_0^6 {\sigma_0^*}^2 \iiint_{-\infty}^{+\infty} \frac{d\beta_1}{2\pi} \frac{d\beta_2}{2\pi} \frac{d\beta_3}{2\pi} \frac{2\pi\delta\left(E(\beta_1) + E(\beta_2) - E(\beta_3)\right)}{8\cosh\frac{E(\beta_1)}{2T} \cosh\frac{E(\beta_2)}{2T} \cosh\frac{E(\beta_3)}{2T}} \coth^2\left(\frac{\beta_1 - \beta_3}{2}\right). \tag{15}$$

The low-T asymptotics of this expression is

$$\left(\frac{1}{T_1}\right)_{q=\pi} \sim \frac{4\sqrt{3}}{\pi}\left(\frac{\sigma_0^6\sigma_0^{*2}}{\Delta}\right)\left(\frac{T}{\Delta}\right)e^{-\frac{2\Delta}{T}}. \tag{16}$$

Putting in the numbers,

$$\left(\frac{1}{T_1}\right)_{q=\pi} \sim 33.54\left(\frac{T}{\Delta}\right)e^{-\frac{2\Delta}{T}} \tag{17}$$

(with the normalization (9), $(1/T_1)_{q=\pi}$ is dimensionless).

(ii) In the high-temperature limit $T \gg \Delta^*$, the quantity $(1/T_1)_{q=\pi}$ is determined by the short-distance asymptotics of the correlation function

$$\langle \tilde{n}_z^-(x)\tilde{n}_z^-(0)\rangle \sim \frac{1}{x}. \tag{18}$$

In this case Sachdev's result [13] gives

$$(1/T_1)_{q=\pi} = \pi \approx 3.1416. \tag{19}$$

(iii) In this limit, we replace the operator σ^* by its zero-temperature expectation value, while pretending that the three remaining operators in (8) are massless. Then

$$\langle \tilde{n}_z^-(x)\tilde{n}_z^-(0)\rangle \sim \frac{\sigma_0^{*2}}{x^{3/4}}. \tag{20}$$

To handle this case, we redo the calculation of [13] for arbitrary exponent η in

$$\langle \tilde{n}_z^-(x)\tilde{n}_z^-(0)\rangle \sim \frac{D}{x^{\eta}}. \tag{21}$$

The magnetic succeptibility is then given by [17]

$$\chi(\omega,k) = \frac{\pi D}{(2\pi T)^{2-\eta}}\frac{\Gamma\left(1-\frac{\eta}{2}\right)}{\Gamma\left(\frac{\eta}{2}\right)}\frac{\Gamma\left(\frac{\eta}{4}-i\frac{\omega+k}{4\pi T}\right)\Gamma\left(\frac{\eta}{4}-i\frac{\omega-k}{4\pi T}\right)}{\Gamma\left(1-\frac{\eta}{4}-i\frac{\omega+k}{4\pi T}\right)\Gamma\left(1-\frac{\eta}{4}-i\frac{\omega-k}{4\pi T}\right)}, \tag{22}$$

which leads to

$$\left(\frac{1}{T_1}\right)_{q=\pi} = \lim_{\omega\to 0}\int_{-\infty}^{+\infty}\frac{dk}{2\pi}\frac{2T}{\omega}\mathrm{Im}\chi(\omega,k) = D\frac{\Gamma^2\left(\frac{\eta}{2}\right)}{\Gamma(\eta)}(2\pi T)^{\eta-1}. \tag{23}$$

In our case ($\eta = 3/4$, $D = \sigma_0^{*2}$),

$$\left(\frac{1}{T_1}\right)_{q=\pi} = \sigma_0^{*2}\frac{\Gamma^2\left(\frac{3}{8}\right)}{\Gamma\left(\frac{3}{4}\right)}(2\pi T)^{-1/4}. \tag{24}$$

Numerically,

$$\left(\frac{1}{T_1}\right)_{q=\pi} = 7.028\left(\frac{T}{\Delta}\right)^{-1/4}. \tag{25}$$

The three temperature dependences (17), (19), and (25) are plotted in Fig. 2.

The mismatch of the asymptotics (ii) and (iii) at $T \approx 3\Delta$ is due to the roughness of the approximation $\langle\sigma^*(t)\sigma^*(0)\rangle = \sigma_0^{*2}$ made in (20). At $T \approx \Delta^*$, this correlation function decays at time scale of order T^{-1} (the same as the triplet-channel correlation functions), which substantially decreases $(1/T_1)_{q=\pi}$. Although for $\Delta^* = 3\Delta$ the limit (iii) is never realized, it clearly indicates that at high temperature $(1/T_1)_{q=\pi}$ slowly decreases with temperature, approaching the constant value of the asymptotics (ii).

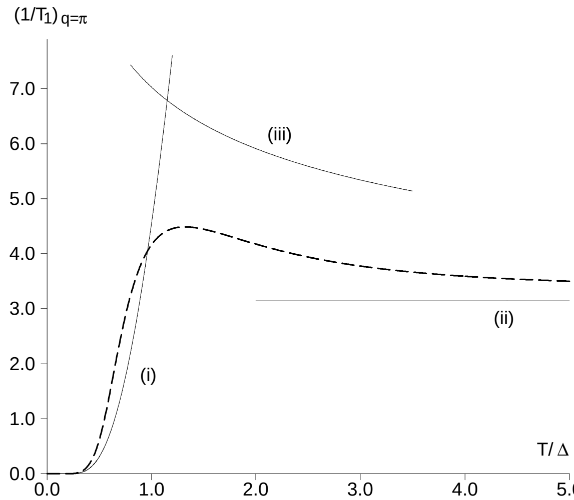

FIG. 2. The three limiting asymptotics of the temperature dependence of $(1/T_1)_{q=\pi}$. Dashed line is the qualitative interpolation of the actual temerature dependence.

Another qualitative consequence of our discussion is the crossover from increasing activation behavior of asymptotics (i) to slowly decreasing high-temperature asymptotics (iii)–(ii). The crossover occurs at $T \approx \Delta$, in spite of the gap 2Δ of the low-temperature asymptotics. This agrees with the experimental results of T. Imai et al., who observed a sharp corssover in $^{63}(1/T_1)$ at $T \approx 425$K in the undoped compound $La_6Ca_8Cu_{24}O_{41}$ [1]. Moreover, the form of the temperature dependence of $^{63}(1/T_1)$ in the hole-doped compounds $Sr_{14}Cu_{24}O_{41}$ and $Sr_{11}Ca_3Cu_{24}O_{41}$ appear qualitatively very close to the proposed form of $(1/T_1)_{q=\pi}$ (including a slowly-decreasing high-T behavior). We may speculate that this can possibly be explained by a different hyperfine coupling of the ^{63}Cu spin in these compounds making $(1/T_1)_{q=\pi}$ contribution dominate in $^{63}(1/T_1)$. On the other hand, hole doping may modify the temperature dependence of $(1/T_1)_{q=\pi}$, which deserves further theoretical study.

It is worth mentioning that our interpolation of $(1/T_1)_{q=\pi}$ (dashed line in Fig. 2) is higher than the asympthotics (i) at low temperature. This comes from the fact that all terms in the low-temperature expansion (10) are positive, and including only the leading term underestimates $(1/T_1)_{q=\pi}$ at low temperatures. This observation ensures that the crossover from (i) to (iii)-(ii) is sufficiently sharp.

To experimentally separate $(1/T_1)_{q=\pi}$ and $(1/T_1)_{q=0}$ contributions in the total spin-lattice relaxation rate $^{63}(1/T_1)$, one can use the fact that $q = \pi$ contribution does not have a singular dependence on the external field. While $(1/T_1)_{q=0}$ singularly diverges at low magnetic fields (as $\log H$ in the free-magnon model [5] or as $H^{-1/2}$ in the spin-diffusion model [3,18]), $(1/T_1)_{q=\pi}$ only weakly depends on the magnitude of the magnetic field.

We hope that further experimental and numerical works will provide a better understanding of the crossover in $(1/T_1)_{q=\pi}$ discussed in this paper.

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